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Study on Relatively High Temperature Superconductors in Layered Pnictide-Oxide Compounds Sm Fe_{1-x}Co_xAsO Measured by Low temperature X-ray diffraction

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ABSTRACT: The parent compound SmFeAsO shows structural phase transition from tetragonal to orthorhombic at the temperature T_D 144K. In the compound SmFe_{1-x}Co_xAsO, with increasing x the structural transition temperature T_D and SDWT_N decrease and reach 0 K at the critical concentration x_c . In our present paper, we report the investigated structural transition temperature, measured by the low temperature X-ray diffraction technique. We have observed that (400) peak of the parent compound SmFeAsO splits into two peaks (400) and (040) spectrum, when the temperature is lower than the structural transition temperature T_D . The structural phase transition of Co doped compounds SmFe_{1-x}Co_xAsO has been studied for x=0, 0.01, 0.025 and 0.075. We have observed that T_D for SmFe_{0.99}Co_{0.01}AsO, SmFe_{0.975}Co_{0.025}AsO and SmFe_{0.925}Co_{0.075}AsO are134K, 118K and 4 K respectively. We determined the phase diagram of T_D versus lattice spacing along c-axis. We have observed that the structural transition decreases with shrinking lattice constant of c-axis like as the decreasing of the structural phase transition temperature with increasing the Co-concentration. Together with the data of the fluorine-doped system Sm(O_{1-x}F_x)FeAs, a phase diagram is drown and is revealed to be similar to that of the Co-doped SmFe_{1-x}Co_xAsO. Also we analyzed the temperature dependence Full Width at Half Maxima (FWHM) and the temperature dependence Area of the profiles for the parent compounds SmFeAsO. The phase diagrams show the similar nature.

Keywords: SmFeAsO, Super conductor, X-ray

I. INTRODUCTION

The High temperature superconductor is usually know as cuprate superconductors. Cuprate loosel refers to a material that can be viewed as containing copper anions. The terms is mainly used in three contexets-oxide materials, anionic coordination complexes and anionic organo copper compounds. Interest in cuprates sharply increased in 1986. After discovery Lanthanum barium copper oxide $La_{2-x} Ba_x CuO_4$, T_c for this material was 35 K. From 1986 to 2008, many cuprate superconductors were identified. The maximum high T_c cuprate superconductors with T_c =127 K observed in $Ba_2Ca_2Cu_3O_{8+x}$ with ambient- $T1_2Ba_2Ca_2Cu_3O_{10}$ in 1988. $T_c = 135$ K achieved in 1993 with the layered cuprate Hg pressure. Studies show that since the discovery of cuprate superconductors, relatively high temperature superconductors in layered pnictide-oxide compounds LnFePnO (Ln = lanthanides, Pn = P, As) in 23 February, 2008, has drawn a lot of interest, when the new class of compounds ReFeAsO (Re = Sm, Nd, Pr, Ce, La) have been prepared [1]. The ReFeAsO system is itself a non-superconducting material. The structural phase transition and antiferromagnetic (AFM) ordering associated with Fe ions in all FeAs-based parent compounds lies in the temperature rang of 100-200 K [2-4]. The low temperature X-ray diffrection study on ReFeAsO (Re = La, Sm, Gd and Tb) observed that structural phase transition of these parent compounds driven magnetically. In this system both c-axis and a-axis lattice constant decreses significantly as Re changes from the bigger iron radious to smaller radious and the structural phase transition temperature also decreases monotonously as the irons radious decreases [5].

Various chemical doping approaches can suppress the structural transition and AFM order and consequently high T_c superconductor appears. The maximum T_c of fluorine-doped iron-based layered La[$O_{1-x}F_x$]FeAs (x=0.05 - 0.12) family is 26 K and enhanchd up to 43 K under 4 Gpa [6]. The unit cell of these compounds contain two molecules, represented by the chemical formula (La_2O_2) (Fe_2O_2). The Fe_2As_2 layere sandwiched between the La_2O_2 layere. Fe_2As_2 acts as a conducting layere and La_2O_2 insulating layers. Replacing O^2 with F the parents compound becomes superconductivity. With the help of the fluorine-doped the maximum T $_c$ of Sm[$O_{1-x}F_x$]FeAs family (x = 0.02) and Ce[$O_{1-x}F_x$]FeAs are 54 K and 41 K respectively [7-8]. Room temperature XRD studies of these compounds shows that with increasing F^- both a-axis and c-axis decreases monoyonically. But F $^-$ doping is very difficult particularly in GdFeAsO system [11-12]. By Thorium (Th^{+4}) substitution for Gd^{+3} in GdFeAsO the superconductivity obserbed at 56 K. The Th-doping expand the lattice with in ab-planes, but shrinking occur along c-axis[13]. The superconductivity also occure in ReFeAs O_{1-x} (Re = Sm, Nd, Pr) by oxygen deficiency in ReO layer instead of F^- doping [14-15]. However, the theoritical calculation for the electron density by G.xu etal [16] suggests that substitution of cobalt for iron is expected to induce electron directly into FeAs layers. Therfore, Co-doping in the FeAs planes is interesting, which would give us importent clues to understand the mechanisms on iron-based superconductor.

Layered crystal structure of Re Fe_{1-x} Co_x AsO is made sandwich FeAs layer with ReO layers. The crystal structure of this system is ZrCuSiAs type. In these system carriers are directly doped with in the FeAs layer by substitution of Co in the tri-valance Co $^{3+}$ state at the place of Fe $^{2+}$. With increasing Co concentration, the structural phase transition temperature (T $_D$) and AFM ordering temperature (T $_N$) decrease and reach 0 K at the critical concentration. The maximum value of T_c occurs near the critical concentration T_c 0 suggesting the QCP which we have already reported [17]. A

domelike $T_c(x)$ curves are widely observed in these iron-based prictides, i.e., Co-dopped $SmFe_{1-x}Co_xAsO_x$ system maximum $T_c = 17.2$ K [18].

 $LaFe_{1-x}Co_xAsO_x$ and

In this paper, we report the investigation of the structural phase transition with increasing Co concentration in SmFe_{1-x}Co_xAsO measured by low-temperature X-ray diffraction. X-ray scattering studies on the ReFeAsO (R=La, Sm, Gd and Tb) samples at room temperature; have found that, as Re is changed from La through Sm and Gd to Tb, both the c-axis and a-axis lattice constant decreases significantly. That is they have observed that as the ions radius decreases the lattice constant also decreases. Also the low temperature X-ray diffraction study on these samples shows that the structural phase transition temperature also decreases monotonously as the ions radius decreases[5]. The study on the polycrystalline samples of LFe_{1-x}Co_xAsO (L=La and Sm) at room temperature shows that Co doping causes the shrinkage of the c axis significantly, while the a-axis nearly unchanged [18-20]. Also shrinkage of the c-axis occurs by the substitution of F in La $[O_{1-x}F_x]$ FeAs and Sm $[O_{1-x}F_x]$ FeAs [1,10]. Similarly, we observed that structural phase transition decreases with decreasing the lattice constant along the c-axis. It should be noted here that we compare our experimental results with F doped Sm $[O_{1,x}F_x]$ FeAs families. It shows that the phase diagram is very similar between the two systems.

We also report the temperature dependence Full Width at Half Maximum (FWHM) and the temperature dependence of Area for those profile of the parents compound SmFeAsO, which are not split below the transition temperature. Our observation shows that increasing FWHM and Area with decreasing temperature below the transition temperature. This paper is organized as follows. In section II, we give experimental works .In section III, we have examined SmFeAsO with different parent compounds. In section IV, we have discussed the results of previous section. Section V is devoted to some remarks.

II. **EXPERIMENTAL WORKS**

2.1. SmFeAsO (parent compound x = 0)

After synthesis the X-ray diffraction at 300K, displayed that the crystal has tetragonal structure with P4/nmm space group having lattice parameters a=0.393726 nm and c=0.849808 nm[18]. Profiles of the θ -2 θ scans of the (400) reflection of the tetragonal unit cell of SmFeAsOat 200 K (T>T_D) and at 130 K, 100 K, 50 K (T<T_D) are shown in Fig.1.(a). These profiles are measured by cryo-cooler with circulating 4He gas. Profiles of the θ -20scans of the (40reflection at 200 K (T>T_D) and at 120 K, 100 K, 50 K (T<T_D) are shown in Fig.1.(b). these profiles are measured by ³He-⁴He dilution refrigerator. From these results we have observed that below T_D the (400) peak splits into two peaks (400) and (040)Spectrums when the temperature is lower than the structural phase transition temperature. These profiles contain $Cu_{\alpha 2}$ and the smaller peak, existing in the right shoulder of the peak, is due to the reflection of the K_{β} line.

We have reported the temperature dependence of the (220) peakd value for SmFeAsO in ref [5]. The temperature dependence of the d value for this sample shows that the structural phase transition was about 144 K [5].

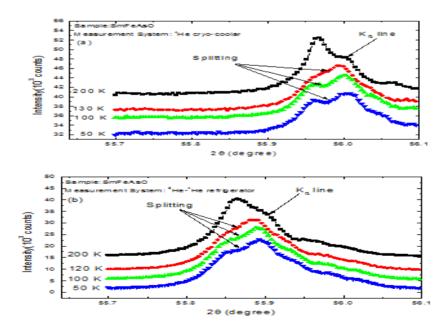


Figure 1: (a) Profiles of the θ -2 θ scans of the (400) reflection of the SmFeAsO above and below the transition temperature measured by 4 He cryo-cooler. (b) Profiles of the θ -2 θ scans of the (400) reflection of the SmFeAsO above and below the transition temperature measured by ³He-⁴He dilution refrigerator.

2.2. $SmFe_{0.99}Co_{0.01}AsO(x = 0.01)$

Profiles of the θ -2 θ scans of the (400) reflection of the tetragonal unit cell of SmFe_{0.99}Co_{0.01}AsO at 200 K (T>T_D) and at 100 K and 50 K (T<T_D) are shown in Fig. 2.(a). These profiles are measured by cryo-cooler with circulating 4He gas. Profiles of the θ-2θ scans of the (400) reflection at 200 K (T>T_D) and at 100 K and 50 K (T<T_D) are shown in Fig.2.(b). These profiles are measured by ³He-⁴He dilution refrigerator. From these profiles we have observed that the crystal

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distortion occurs below the transition temperature. The temperature dependence of the d value for this sample will be published in the near future. Our experimental results show that the crystal distortion from the tetragonal to orthorhombic occurs at 134 K.

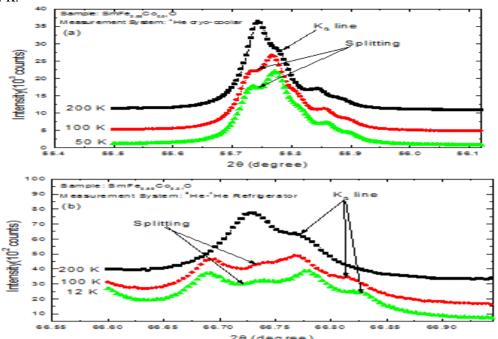


Figure 2:(a) Profiles of the θ -2 θ scans of the (400) reflection of the SmFe_{0.99}Co_{0.01}AsO above and below the transition temperature measured by 4 He cryo-cooler. (b) Profiles of the θ -2 θ scans of the (400) reflection of the SmFe_{0.99}Co_{0.01}AsO above and below the transition temperature measured by 3 He- 4 He dilution refrigerator.

2.3. $SmFe_{0.975}Co_{0.025}AsO(x = 0.025)$

Similarly, the Profiles of the θ -2 θ scans of the (400) reflection of the tetragonal unit cell of SmFe0.99Co0.01AsO at 150 K (T>T_D) and at 50 K and 12 K (T<T_D) are shown in Fig. 3.(a). Profile of the θ -2 θ scans of the (400) reflection at 150 K (T>T_D) and at 50 K and 12 K (T<T_D) are shown in Fig.3.(b). From these profiles we have observed that the crystal distortion occurs below the transition temperature. Our experimental results show that the crystal distortion from the tetragonal to orthorhombic occurs at 118 K.

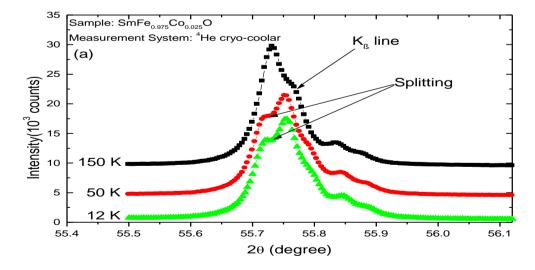


Figure 3:(a) Profiles of the θ -2 θ scans of the (400) reflection of the for SmFe_{0.975}Co_{0.025}AsO above and below the transition temperature measured by ⁴He cryo-cooler.

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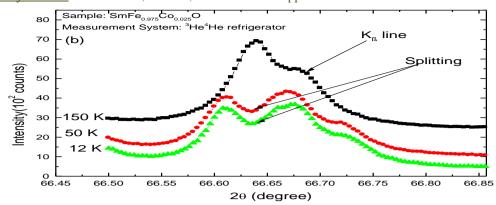


Figure 3:(b) Profiles of the θ -2 θ scans of the (400) reflection of the for SmFe0.975Co0.025AsO above and below the transition temperature measured by ${}^{3}\text{He}{}^{-4}\text{He}$ dilution refrigerator.

III. EXPERIMENTAL RESULTS

We have explained structural phase transition temperature T_D for SmFeAsO and for $SmFe_{1-x}Co_xAsO$ samples with x=0, 0.01, 0.025 & 0.075. The room temperature X-ray diffraction (XRD) pattern for $SmFe_{1-x}Co_xAsO$ samples with x=0, 0.01, 0.025 & 0.075 along with their Rietveld analysis was shown that the structure of all compositions of x was the tetragonal space group P4/nmm, but they changed there structure from tetragonal to orthorhombic on cooling. We analyzed above expression by measuring profiles of the θ -2 θ scans using cryo cooler with circulating ⁴He gas and ³He-⁴He dilution refrigerator. By observation, we found that for x=0.01 in $SmFe_{1-x}Co_xAsO$ T_D near about 134K.

IV. DISCUSSIONS

The structural characterization of $SmFe_{I-x}Co_xAsO$ samples was reported by ref. [18]. They have observed that as the Co doping causes the shrinkage of the c-axis significantly, while the a-axis remains nearly unchanged. The unchanged a-axis suggests no significant oxygen deficiency present in our samples. The superconductivity also observed in oxygen-deficient ReFeAsO_{1-x}, oxygen-deficient in Sr-doped LaFeAsO[14, 21], the F^- doping at the oxygen site in Re $[O_{1-x}F_x]$ FeAs (Re = La and Sm) and also F^- doped in Ce $[O_{1-x}F_x]$ FeAs. The entire reported superconductors showed remarkable change in both a-axis and c-axis [1, 8, and 9]. Since a-axis remains unchanged, so in Co doped compound Superconductivity occurs due to the density of sate in the FeAs layers which are increased by the Co doping. Shrinking of the c-axis observed up to x = 0.3[18, 20]. The shrinkage of c-axis occurs, because Co^{2+} ions radius is smaller as compared with Fe²⁺. This suggests the strengthening of interlayer Coulomb attraction, implying the increase in density of negative charge in FeAs layers by Co doping.

The antiferromagnetic ordering in the parent compound originated from the nearest-neighboring interactions, bridged by As 4p orbital's [22-24]. These interactions are anti-ferromagnetic, which makes frustrated magnetic ground state. Doping Co into the Fe site, interaction between Co and Fe atoms distort the AFM order. The superconductivity occurs in $SmFe_{1-x}Co_xAsO$ at x = 0.05[17-18] suggest that Co doping at Fe site play an important rule to suppressed structural phase transition and spin density wave. We are interested to know how can the structural phase transition temperature is related to the c-axis. For this purpose we have plotted the transition temperature against the lattice constant of the c axis. The values of the c- axis with different Co concentrations were taken from ref. [18].

Fromour experimental result we have observed that T_D for SmFe_{0.99}Co_{0.01}AsO, SmFe_{0.975}Co_{0.025}AsO and SmFe_{0.925}Co_{0.075}AsO are 134 K, 118 K and 4 K respectively. Fig. 4.(a). Shows the structural phase transition temperature vs c-axis curve.

This figure clearly shows that the structural transition temperature is strongly related to the c-axis lattice constant. Finally, we plot a curve for comparing our experimental results with F-doped Sm[O_{1-x}F_x]FeAs family as shown in fig.4.(b). Data are collected from S. Margadonnal et al. [10]. They have studied crystal structure phase transition of Sm [O_{1-x}F_x] FeAs with x = 0, 0.05, 0.10, 0.12, 0.15 and 0.20. The structural phase transition observed up to x = 0.12 and T_D approximately 50 K. On the other hand, our experimental studies on $SmFe_{1-x}Co_xAsO$ is with x = 0, 0.01, 0.25 and 0.075. The lowest temperature of the structural phase transition was about 4 K with x = 0.075.

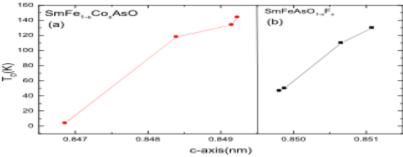


Figure 4: Structural phase transition vs c-axis curve.

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Experimental findings are listed in Table. 1.

Table. 1.

System	$Sm[O_{1-x}F_x]FeAs$	$SmFe_{1-x}Co_xAsO$
Structuralphase transition	$0 \le x \le 0.12$	$0 \le x \le 0.075$
region.		
Lowest T_D	Approximately 50K	4 K
Schering c-axis region	$x \le 0.20$	$x \le 0.30$

The Co doped $SmFe_{1-x}Co_xAsO$ system shows both similarity and differences in comparison with F^- doped $Sm[O_1]$. _xF_x|FeAs system. The similarities are: (1).The structural phase transition in both system decreases with increasing the concentration. (2). The phase diagrams shown in fig.4. look very similar. The differences between the two systems are listed in Table. 1. Here, we noted the following points: (1) The Co doping destroys the AFM, SDW order more strongly. (2). The T_D is significantly lowered in Co-doped system. (3). In the Co doped system the chemical phase boundary region is longer. The first and the second can be understood in terms of the variation in exchange interactions. Upon doping Co into Fe site, the original AFM super exchange interactions may be changed into a double exchange between Co and Fe atom, which destroys AFM order. Different number of 3d electrons at Fe²⁺ and Co²⁺ validates the double exchange in the form of, $Fe^{2+}3d \leftrightarrow$ $As^{3-}4p \leftrightarrow Co^{2+}3d$ and logous to the classic double exchange interaction in perovskite-type manganite's [25]. Due to the double coupling in the Co doped system, the structural phase transition driven to the lower temperature with x = 0.075, which is much smaller than the F-doped system. Although at room temperature c-axis shrinkage up to x = 0.30 in the Co doped system, but T_D of this compound sharply driven to 4 K with x = 0.075. On the other hand at x = 0.03, superconductivity does notappear in this compound [20], due to the disorder effect in (Fe/Co) as layers. In the F^- doped $Sm[O_{1-x}F_x]FeAs$, c-axis shrinkage up to x=0.02 and themaximum superconductivity occurs at 0.02 concentration [7]. It is the temperature dependence of F ullWidth at Half Maximum (FWHM) and the temperature dependence of Areas of those peaks, which are not splitted below the transition temperature. The profile of the θ -2 θ scans of the (220) reflection of the tetragonal unit cell of SmFeCoAsOat 150 K (T>T_D) and at 132 K and 130 K (T<T_D) are shown in Fig.5.

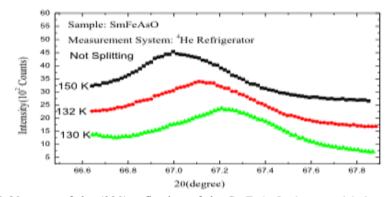


Figure 5: Profile of the θ -2 θ scans of the (220) reflection of the SmFeAsO above and below the transition temperature measured by ⁴He cryo-cooler.

In Fig.6(a) FWHM of reflection is plotted agains temperature and Fig.6(b) is the Enlargement of Fig.6(a) in the transition temperature region.

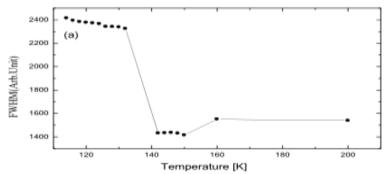
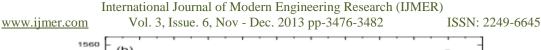


Figure 6:(a). FWHM vs Temperature curve for (220) reflection of SmFeAsO.



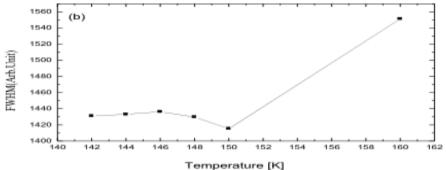


Figure 6:(b). The Enlargement of fig. 6(a) in the Transition Temperature region.

Figure. 6.(a) and 6(b) show the softening down to about 148K, with decreasing temperature. Suggesting the occurence of the crystal distortion. It also makes a plateau around the transition temperature. At the lowest temperature region it is nearly saturated. Finally, our observation shows that FWHM increases below the transition temperature. Figure. 7(a) shows the temperature dependence of Area of (200) reflections and Fig. 7(b) is the Enlargement of Fig. 7(a) in the transition temperature region.

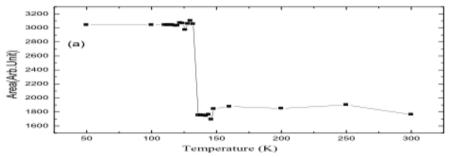


Figure 7:(a). Area vs Temperature curve for (220) reflection of SmFeAsO.

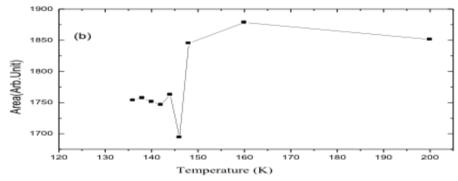


Figure 7:(b). The Enlargement of fig.7(a) in the Transition Temperature region.

It also shows that softening occurs at about 150K. Also it start to increase with decreasing temperature. The temperature dependence of Area curve also shows the plateau around the transition temperature and also in the lowest temperature region it becomes saturated. Our observation shows that area of the peak increases below the transition temperature. The phase diagrams of temperature dependence FWHM and Area show a very similar nature.

V. CONCLUSION

From our experimental result we have observed that the transition temperatures T_D for SmFeAsO, SmFe_{0.99}Co_{0.01}AsO, SmFe_{0.975}Co_{0.025}AsO and SmFe_{0.925}Co_{0.075}AsO are 144 K, 134 K, 118 K and 4 K respectively. Our experimental results clearly show that as the Co concentration increases the structural phase transition temperature decreases like as shrinkage of the c axis lattice constant. That is structural transition temperature is strongly related to the c-axis lattice constant. In comparison to its F^- doped Sm[O_{1-x}F_x]FeAs system the phase diagram shows the similarity, but in the Co-doped system the structural phase transition driven to more lower temperature with small concentration compare to F^- doped Sm[O_{1-x}F_x]FeAs system.We have observed that the structural transition decreases with shrinking lattice constant of c-axis like as the decreasing of the structural phase transition temperature with increasing the Co concentration. Also we observed increasing Full Width at Half Maximum (FWHM) and Area with decreasing temperature below the transition temperature.

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